

REMARKS

Re-examination and reconsideration of the subject matter identified in caption, pursuant to and consistent with 37 C.F.R. §1.111, and in light of the remarks which follow, are respectfully requested.

Claims 39-47, 52-54, 56-63, 66, 67 and 69-76 remain pending in this application.

Applicants acknowledge with appreciation the indication that claims 59-62, 66, 67 and 69-76 are allowed.

Claims 39-47 and 63 were rejected under 35 U.S.C. §103(a) as obvious over newly-cited U.S. Patent No. 4,044,171 (Müller et al '171) for the reasons given in paragraphs (3) and (4) of the Office Action. Reconsideration and withdrawal of this rejection are respectfully requested for at least the reasons which follow.

The Office Action refers to the statement in column 3, lines 31-36 of the reference that dimerization by heating in the absence of a catalyst is most suitable at temperatures between 120° and 150°C. However, this passage does not indicate what type of isocyanate may be dimerized under these conditions.

The Office Action also refers to column 4, lines 20 *et seq.* Again, there is no disclosure therein of which, if any, of the listed diisocyanates may be dimerized in the absence of a catalyst. The procedures for preparing dimerized diisocyanates in the working Examples all employ a catalyst and an aromatic diisocyanate (Examples 1-6, 20 21). Dimeric hexamethylenediisocyanate is mentioned in Examples 7, 8 and 23, but the method of preparation is not disclosed.

Thus, while Müller et al '171 generally mentions diisocyanate dimerization, there are no specific details of preparing dimers by heating isocyanate monomers having NCO groups borne by sp^3 carbon atoms in the absence of a catalyst.

Müller et al '171 refers to the publication "Polyurethanes: Chemistry and Technology, Part I," Saunders – Frisch, pages 91-94 (*see* column 2, lines 63-65). On page 91 of the Publication, the authors state: "Dimers of aliphatic isocyanates have not been obtained; trimerization occurs instead" (lines 4-5 from the bottom of the page). A copy of the relevant pages is attached for the convenience of the Examiner.

Moreover, when a catalyst is employed, Müller et al '171 disclose that "the aromatic or mixed aromatic-aliphatic uretdione diisocyanates crystallize from the reaction in the course of dimerization" (column 3, lines 49-51). This disclosure teaches away from the presently claimed invention where low viscosity (i.e., fluid, not crystals) (poly)isocyanate compositions are obtained.

Respectfully, Müller et al '171 contains no explicit teaching that would have motivated those of ordinary skill in this art to prepare low viscosity (poly)isocyanate compositions containing uretdione dimers by heating an isocyanate monomer having an isocyanate group borne by sp^3 carbon atoms in the absence of a catalyst at a temperature of 50°-200°C for not more than 24 hours. Those of ordinary skill in this art would not be led by the disclosure of this reference to the presently claimed invention.

In view of the above, the §103 rejection over Müller et al '171 should be withdrawn. Such action is earnestly solicited.

Claims 52-54 and 56-58 were rejected under 35 U.S.C. §103(a) as obvious over newly-cited U.S. Patent No. 5,369,207 (Wolff et al '207) in view of Müller et al '171 for the reasons given in paragraphs (5) and (6) of the Office Action. Reconsideration and withdrawal of this rejection are requested for at least the following reasons.

Wolff et al '207 is directed to the preparation of isocyanurate and urethane group-containing polyisocyanate mixtures which optionally may contain "small amounts of

uretdione groups." Note column 6, lines 29-30. The process of this reference performs a (cyclo)trimerization of a diisocyanate followed by reaction with alcohols. There is no disclosure of any reaction conditions for the preparation of mixtures containing uretdione groups. The viscosities disclosed in Table I widely range from 420 to 1240 mPa.s at 25°C. Example 10 discloses that the mixture is cloudy which strongly suggests that some crystallization has occurred.

Even if one sought to combine the respective disclosures of the cited documents, there would have been no suggestion of the present invention. As discussed above, Müller et al '171 does not disclose the dimerization of aliphatic isocyanates by heating in the absence of catalyst at 50° - 200°C, nor does Müller et al '171 disclose the preparation of low viscosity (poly)isocyanate compositions. Wolff et al '207 does not disclose the preparation of uretdiones from aliphatic isocyanates using any process. Modifying the invention disclosed in Wolff et al '207 in accordance with Müller et al '171 would not lead to the presently claimed invention.

For at least these reasons, the §103(a) rejection based on Wolff et al '207 in view of Müller et al '171 should be reconsidered and withdrawn. Such action is earnestly solicited.

From the foregoing, further and favorable action in the form of a Notice of Allowance is believed to be next in order and such action is earnestly solicited. If there are any questions concerning this paper or the application in general, the Examiner is invited to telephone the undersigned at (703) 838-6683 at his earliest convenience.

Respectfully submitted,

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Part I

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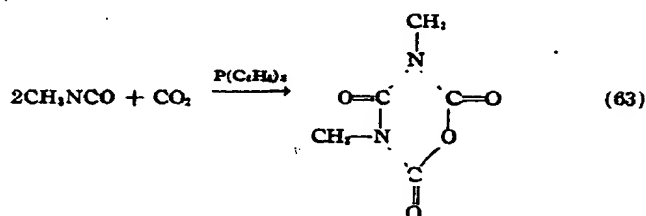
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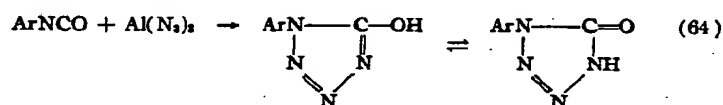
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Slotta and Tschesche (221) reported that methyl isocyanate condensed with carbon dioxide as in reaction (63). This is a very rare



observation of carbon dioxide entering into a reaction with the isocyanate, rather than being evolved from such a reaction. No reaction was obtained, however, with carbon oxysulfide, carbon disulfide, phenyl isothiocyanate, acetonitrile, nitromethane, quinone, acetylene, ethylene, or amylene under the influence of triethylphosphine. With aldehydes, aldoketenes, carbon suboxide, and cyanic acid in the presence of triethylphosphine, only polymers of these substances and the trimer of methyl isocyanate were obtained. No condensation occurred with either dimethylketene or diphenylketene. Treatment with diazomethane gave an insoluble, amorphous substance which could not be identified.

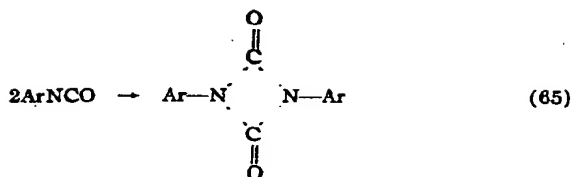
Aluminum azide reacts with aromatic isocyanates (eq. 64) to give a cyclic structure that may be considered as a product of addition to the C—N double bond (113).



7. Dimerization

As a special case of the reaction with unsaturated compounds, aromatic isocyanates may react with themselves to form dimers, "uretidine diones" (eq. 65). The reaction also is analogous to the dimerization of ketenes. Dimers of aliphatic isocyanates have not been obtained; trimerization occurs instead.

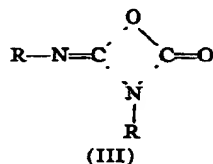
Dimerization is catalyzed vigorously by trialkylphosphines (31,84, 109,110), and more mildly by tertiary amines such as pyridine (152, 192,225). Certain aromatic isocyanates such as 4,4'-diphenylmethane



diisocyanate dimerize slowly on standing, even without catalysts. The rate of dimerization of many other isocyanates is greatly retarded if an *ortho* substituent is present.

Molecular weight measurements of phenyl isocyanate dimer are in agreement with the dimer structure. These measurements were based on ebullioscopic methods and observed molecular weights were somewhat lower than theoretical, suggesting some dissociation in the boiling solutions (31). The crystal structure has also been determined (41), and is in agreement with the symmetrical structure as indicated.

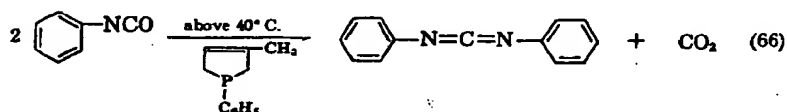
Although the diketo structure as has been shown is generally accepted as being correct, some consideration has been given to an alternate structure (III) (88). The bulk of the evidence is in favor of the symmetrical diketo form, however.



When pyridine is used as a catalyst for dimer formation, it is most conveniently applied as the solvent medium for the reaction (225). Phosphine catalysts are much more active than pyridine and are used only in trace amounts. A convenient method of controlling phosphine-catalyzed dimerizations (21) involves the addition to the reaction mass of an alkylating agent such as benzyl chloride in an amount stoichiometrically equivalent to the substituted phosphine present. Complete deactivation of the catalyst results.

Among the phosphine catalysts the trialkyl derivatives are the most active. Dialkylaryl compounds, of which dimethylphenylphosphine is a good example, are more convenient in practice, however, because their action is more moderate, permitting better control of the exothermic dimerization reaction (227). When the phosphorus atom

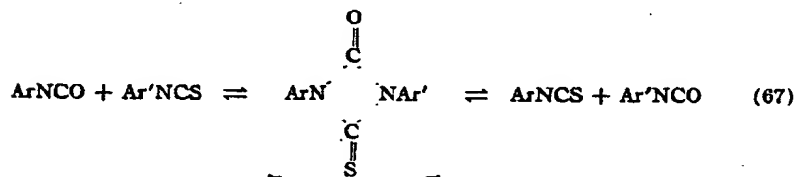
carries two phenyl groups, its efficiency as a catalyst is reduced, and triphenylphosphine exhibits no catalytic activity. One member of the phosphine family, 3-methyl-1-phenyl-3-phospholine, deserves special attention because of its unique activity as a catalyst for carbodiimide formation (eq. 66). The dimer is not an intermediate in this



reaction, and the diphenylcarbodiimide is formed in nearly 90% yield. Phenylisocyanate dimer may be obtained using this catalyst only under mild conditions in dilute solution (12).

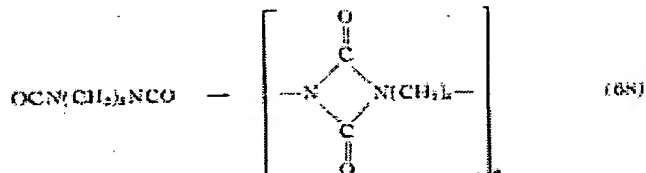
Phenyl isocyanate dimerization is a reversible reaction. The equilibrium mixture is not affected by changes in catalyst concentration. However, the conversion to dimer at equilibrium increases with decreasing temperature, with 71–74% conversion reported at 25°C. and 90% at 10°C. (12). The reaction may be forced in one direction, however, as by the precipitation of dimer from the reaction mixture, thus giving good yields at higher temperatures (216).

"Mixed dimers" have been prepared from two different isocyanates (216), and an analogous combination of α -naphthyl isocyanate and phenyl isothiocyanate apparently has been formed (eq. 67). Four



products, including α -naphthyl isothiocyanate, were isolated after 14 hours of reaction at 215–225°. Formation of the "mixed dimer" as an intermediate would be a logical explanation (52). The formation of a polymeric dimer (eq. 68) by letting trimethylene diisocyanate stand for a week was reported by Iwakura and Ishizuka (121).

The preparation and properties of many dimers of mono- and diisocyanates, as well as mixed dimers of two monoisocyanates, have been reported by Siefken (216), and properties have been summarized by

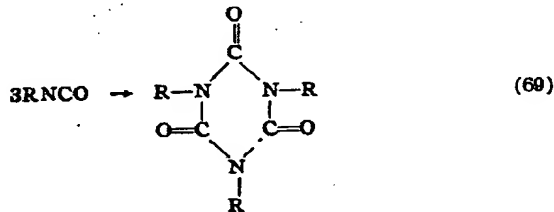


Arnold, Nelson, and Verbanc (12). The chemistry of isocyanate dimers was also reviewed recently by Taub and McGinn (235).

8. Trimerization

Both aliphatic isocyanates and aromatic isocyanates can form trimers. This reaction is another special example of a reaction of an isocyanate with an unsaturated compound. Catalysts such as triethylphosphine, which promote dimerization of aromatic isocyanates, catalyze trimerization of the aliphatic compounds (109). Other catalysts that have been used to induce the trimerization of either aromatic or aliphatic isocyanates include calcium acetate (84), potassium acetate (111), sodium formate (111), sodium carbonate (111), sodium methoxide (155), triethylamine (112), oxalic acid (17), sodium benzoate in dimethylformamide (19), and a large number of soluble compounds of iron, sodium, potassium, magnesium, mercury, nickel, copper, zinc, aluminum, tin, vanadium, titanium, and chromium (107), titanium tetrabutryate (142), and oxygen (134). Friedel-Crafts catalysts are also effective in bringing about polymerization (25). Perhaps because of steric hindrance, *tert*-butyl isocyanate reportedly does not trimerize, even with triethylphosphine (37). Similarly, the presence of *ortho* substituents on an aromatic isocyanate greatly reduces its ease of trimerization.

The generally accepted structure for the trimer is that of a trisubstituted isocyanurate (eq. 69). Phenyl isocyanate trimer of this



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